

Study and evaluation of cesium chemisorption behavior onto steel under simulated nuclear reactor severe accident environment in 400 °C.

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Abstract: Since Cs chemisorption might occur not only within RPV but also within PCV at 1F, Cs chemisorption behavior onto steel was investigated at 400 °C. X-ray diffractometer(XRD) and Raman spectroscopy were used for the identification of Cs compounds formed on carbon steel and stainless steel 304(SS304), which is the structural material of PCV and RPV used in 1F respectively. As a result, CsFeO₂ was found to be the major chemisorption product on both carbon steel and SS304.

Keywords: cesium, chemisorption, steel, severe accident

1. Introduction

Since cesium (Cs) is one of the major radioactive sources, information on the distribution of Cs within reactors is necessary for the fuel debris retrieval at the Fukushima Daiichi Nuclear Power Station (1F). For estimation of the Cs distribution, JAEA has paid attention to the chemisorption of Cs on structural materials such as stainless steels because BWR has a steam separator and dryer that has a large surface area made of SS304. They are located in the upper area of the reactor pressure vessel (RPV), which was experiencing a relatively higher temperature during the accident. However, a recent JAEA study indicates that Cs chemisorption could occur at 400 °C [1] while earlier report indicated Cs chemisorption occurs above 600°C for SS304 [2]. Moreover, cesium ferrate (CsFeO₂) is identified as the major chemisorption species. It has been reported that the Cs chemisorption occurs above 600°C for SS304 [2]. Thus, there is a possibility that Cs might be chemisorbed not only on the superstructure surface area inside the RPV but also on a wider area including the steel surface of the primary containment vessel (PCV) at 1F. Hence, to predict the chemisorbed Cs distribution accurately, this study aims to investigate the chemisorption behavior of Cs onto carbon steel (similar to the PCV structural material).

2. Experimental method

3 different types of steel were used in this study which is carbon steel 480 (SGV), pure iron (Fe) with a purity of 99.5% which is expected to have the same level of Fe chemical activity as that of SGV, and stainless steel type 304 (SS304). The dimension of the sample is 10 mm x 10 mm x 2 mm. Cs chemisorption tests were performed at 400°C in Ar-4%H₂-20%H₂O for SS304 and in Ar-5%H₂-5%H₂O for both SGV and pure Fe. Cs hydroxide monohydrate (CsOH·H₂O) with a purity of 99.95% was used as a Cs vapor source and vaporized at 650°C in the chemisorption tests. Due to most of the Cs compound is highly hygroscopic, a nitrogen-filled glove box with a controlled dew point of -71 to 79 °C was used during sample preparation and storage. Cs compounds formed on the steels were identified by using XRD and Raman spectroscopy. In order to determine amounts of water-soluble and water-insoluble Cs chemisorbed onto the samples, samples after the chemisorption tests were soaked in water at 30°C for 1 h and then soaked in boiling aqua regia for about 10 min. Amounts of water-soluble and water-insoluble Cs were determined by using inductively coupled plasma mass spectrometry (ICP-MS).

3. Results and discussion

XRD analysis of the SGV sample indicated that CsFeO₂ and Cs₂CO₃·3H₂O were presented as shown in Fig.1. While Raman analysis of the SGV sample indicated that CsFeO₂ was homogeneously distributed yet Cs₂CO₃ seemed to be localized. Cs₂CO₃ might be formed by being exposed to air due to the hydrate formed, which can be easily decomposed during the chemisorption test. Fig. 2 shows the determined amounts of water-insoluble (black line) Cs and ratios of amounts of water-insoluble Cs to those of water-soluble Cs (Blue line) for the SGV, SS304, and pure Fe samples. Meanwhile, the amounts of water-insoluble Cs for the SGV samples seem to be smaller than those for the SS304 samples. However, the ratios of water-insoluble Cs amount to those of water-soluble Cs for the SGV samples are higher than those for both SS304 and pure iron samples. Therefore, it is considered that Cs chemisorption for the carbon steel would be easier to occur than that for stainless steel because of the higher chemical activity of Fe.

Reference

[1] 中島他、原子力学会 2021 春の年会 3G02. [2] B. R. Bowsher, et al., AEEW-R 1863, (1990).

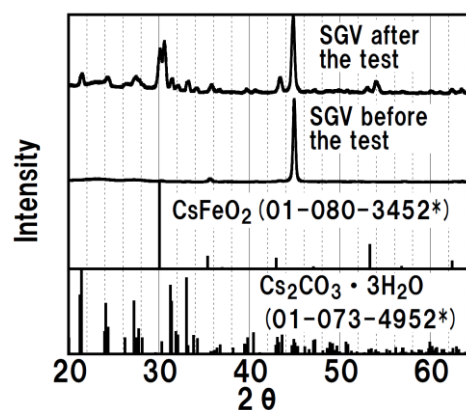


Fig. 1. XRD patterns of SGV before and after chemisorption test at 400 °C (*ICDD pdf number)

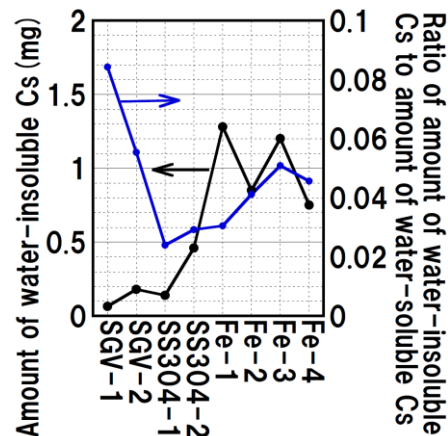


Fig. 2. Amounts of water-soluble and water-insoluble Cs for SGV, SS304 and pure Fe after chemisorption tests at 400 °C